KERNFORSCHUNGSZENTRUM

KARLSRUHE

Separation of Americium and Curium
from Highly Radioactive Waste Solutions
Part I.: Laboratory Batch Studies
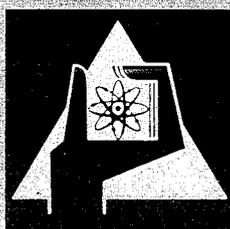
Z. Kolarik, G. Koch, H. H. Küsel, J. Fritsch

KERNFORSCHUNGSZENTRUM KARLSRUHE

Februar 1972 · KFK-1553

INSTITUT FÜR HEISSE CHEMIE

Separation of Americium and Curium from
Highly Radioactive Waste Solutions

Part I.   Laboratory Batch Studies

(PACT-Bericht Nr. 18)

Z. Kolarik, G. Koch, H.H. Küsel and J. Fritsch

Gesellschaft für Kernforschung mbH Karlsruhe

Abstract:

Optimum conditions of a solvent-extraction process for isolating
Am and Cm from highly radioactive wastes have been sought.
Highly extractable multivalent fission and corrosion products
^{95}Zr(IV), ^{106}RuNO(III), Fe(III) etc. can be separated as preci-
pitates in decomposing nitric acid by reduction with formic
acid. Am and Cm can be then extracted together with fission
lanthanides(III) by a solution of di(2-ethylhexyl) phosphoric
acid and tributyl phosphate in an alkane solvent and separated
from the lanthanides(III) by a selective back extraction with
an aqueous solution of diethylenetriaminepentaacetic and lactic
acids. The spent organic phase can be recovered by washing with
a 2 - 5M nitric acid solution and, subsequently, a sodium lactate
and sodium hydroxide solution.

Abstrakt:

Für einen extraktiven Prozeß zur Gewinnung von Am und Cm aus
hochaktiven Abfallösungen wurden optimale Bedingungen untersucht.
Gut extrahierbare Spalt- und Korrosionsprodukte, wie z.B. ^{95}Zr(IV),
^{106}RuNO(III), Fe(III) usw. können bei Zersetzung von Salpeter-
säure durch Ameisensäure als Niederschläge abgetrennt werden.
Am und Cm können dann mit einer Lösung von Di(2-äthylhexyl)-
phosphorsäure und Tributylphosphat in n-Alkan, zusammen mit den
Lanthaniden(III) extrahiert werden. Die Trennung von den
Lanthaniden(III) kann durch eine selektive Rückextraktion mit
einer wäßrigen Lösung von Diäthylentriaminpentaessigsäure und
Milchsäure erfolgen. Zur Regenerierung der gebrauchten organi-
schen Phase werden eine Wäsche mit 2 - 5 M Salpetersäure und
eine darauffolgende Wäsche mit einer Lösung von Natriumlactat
+ Natriumhydroxid vorgeschlagen.

This report describes part of the development of a solvent extraction process applicable for large scale isolation of americium and curium from highly active waste solutions resulting in the reprocessing of spent nuclear fuels. The extractant used in the process is di(2-ethylhexyl) phosphoric acid (henceforth HDEHP) which has been shown to extract trivalent actinides with a good efficiency and, under certain conditions, a good selectivity. The waste solutions contain fission products, metallic corrosion products, sodium nitrate, and large amounts of nitric acid. Thus the process under development consists of the following steps (1):

1) Decomposition of nitric aicd.
2) Extraction of americium, curium and the fission lanthanides, while leaving the bulk of the other fission or corrosion products in the aqueous phase.
3) Selective back extraction (partitioning) of americium and curium, while leaving the fission and corrosion products in the organic phase.
4) Removal of the fission and corrosion prodcuts from the spent organic phase.

The decomposition of nitric aicd has been studied by the Waste Disposal Department (2), and will not be described in detail in this paper. Nitric acid is reduced by formic acid with the formation of gaseous products only, i.e. CO_2, NO_2, NO, and N_2. A pH value as high as 8 can be reached at the end of the denitration reaction. This makes it possible to choose between one of the two following concepts of the extraction process.

Concept No. 1:

The denitration reaction is stopped at such a concentration of nitric acid at which virtually no precipitation of fission or corrosion products occurs. The extraction of interfering multi-

valent elements like Zr(IV), Nb(V), Fe(III), etc. must then
be suppressed by the addition of a complexing agent (for example
lactic or citric acids) which would not prevent the extraction
of Am(III) and Cm(III).

Concept No. 2:

Nitric acid is decomposed to such a pH value at which certain
fission and corrosion products are precipitated in the form
of hydroxides, zirconium molybdate, etc., while Am(III), Cm(III)
and lanthanides(III) remain in the solution. The latter also
contains major amounts of slightly extractable elements (Sr(II),
Ba(II), Cs(I) etc.) but only traces of highly extractable
multivalent elements. No addition of complexing agents to the
aqueous phase may then be needed.

Following our studies of concept No. 1, it was found by workers
of the Waste Disposal Department that the presence of complexing
agents could make difficult the final treatment of the highly
radioactive waste solutions resulting from the Am/Cm separation
process. Thus concept No. 2 was subsequently studied in detail
and was preferred in the later development work.

In this report the search is described for the optimum condit-
ions of an efficient and selective americium separation. Only
the behaviour of Am(III) was studied, that of Cm(III) can be
expected to be very similar both in the presence and absence
of complexing agents. In our concept No 1, the main complexants
in the first (extraction) stage of the process were citric or
lactic acids; both exhibit sufficient complexing power, are well
soluble in water, and their cost is low enough for a large scale
use. Nitrilotriacetic acid (henceforth NTA) was used as the ad-
ditional complexant in combination with citric or lactic acids;
NTA was shown to suppress the extraction of Zr(IV) more strongly
than that of Am(III) (3), and, moreover, it prevents the deposition
of solid lanthanide(III) citrates occuring on longer standing of

the aqueous phase before the extraction (1). A mixture of
C_{10} to C_{13} n-alkanes was utilized as the diluent, because of
the chemical and radiation stability of saturated straight-
chain hydrocarbons. Since the reuse of the HDEHP/n-alkane
extractant is possible only after washing with an alkaline
solution, tri-n-butyl phosphate (henceforth TBP) was added as a
modifier, i.e. an agent preventing the formation of a third
phase during this step.

E X P E R I M E N T A L
_____

Concentrations of fission and corrosion products in the simulat-
ed waste solutions were taken from (2); the concentrations in
the aqueous phase before extraction are given in Table 1.
During preparation of the simulated waste solutions a solid
containing mainly Zr and Mo (most probably zirconium(IV) molyb-
date) is formed. Batch extraction experiments were performed
at room temperature in one-way glass flasks (total volume
12 - 13 ml) closed with polyethylene lids. The phases were
separated by standing and samples of suitable volumes were
taken for measurements of radioactivity and pH. If not other-
wise given, the volumes of the organic and aqueous phases were
3.00 and 2.00 ml respectively in the extraction experiments
(section I), and 2.00 ml each in the experiments on the back
extraction (section II and III). The shaking time was 3 min.
Other experimental details are given in legends of figures and
tables. In experiments with complexing aqueous phases the pH
was measured by the electrode pair glass electrode - S.C.E.
(G 222 B and K 100, both Radiometer, Copenhagen) in connection
with the pH Meter Model 26 (Radiometer Copenhagen). In the
absence of complexing agents in the aqueous phase the pH was
usually determined by an alkalimetric titration. Gamma radio-
activity of ^{241}Am, ^{152,154}Eu, ^{144}Ce, ^{85}Sr, ^{59}Fe and ^{106}Ru +
^{106}Rh was measured by a well-type NaI(Tl) scintillation counter

and that of ^{95}Zr and ^{95}Nb by a Ge-Li detector connected with a 4096 channel analyzer (TMC). HDEHP (Union Carbide), TBP (Merck) and a mixture of n-alkanes (Gelsenberg) were used as received.

RESULTS AND DISCUSSION

I. Behaviour of Am(III) and Fission and Corrosion Products in the Extraction Stage

1. Extraction from Complexing Media (Concept No. 1)

The feed was prepared by mixing a stock solution of simulated fission and corrosion products with stock solutions of lactic or citric acid, NTA, sodium hydroxide, a radioactive tracer, and water in such ratios that the concentrations given in Table 1 and in legends to Figures 1 - 5 and Tables 2 - 5 were adjusted. pH Dependencies of the extraction of Am(III), some fission products, and Fe(III) are shown in Figs. 1 and 2. The separation of Am(III) from Sr(II) is fairly good at pH<2.5 with both complexants, but the distribution ratios of Am(III) in this pH range are by a factor of 5 to 10 higher in the presence of lactic acid than in the presence of citric acid.

In attempt to enhance the distribution ratios of Am(III) in the citric acid system, the NTA concentration was lowered from 0.1 to 0.05 M (Fig. 2). This improved the extraction of Am(III) at pH<2, but not sufficiently. Also the extraction of Eu(III) was augmented at pH<2 by lowering the NTA concentration, while the behaviour of Ce(III) was apparently not affected. This can be ascribed to the higher stability of the Am(III) and Eu(III) chelates of NTA in comparison with the stability of the Ce(III) chelate (4,5). Lowering the citric acid concentration below the level used, namely 0.5 M, would not be reasonable. A considerable

Table 1

Concentrations of fission and corrosion products in the aqueous phase before the extraction (taken from (1)).

Salt	Conc./M	Salt	Conc./M
$RbNO_3$	0.0035	$Zr(NO_3)_4$	0.033
$CsNO_3$	0.014	Na_2MoO_4	0.023
$Sr(NO_3)_2$	0.0085	$UO_2(NO_3)_2$	0.0052
$Ba(NO_3)_2$	0.0066	Na_2TeO_4	0.0014
$Y(NO_3)_3$	0.0043	$Mn(NO_3)_2$	0.0436
$La(NO_3)_3$	0.029	$RuNO(NO_3)_3$	0.010
$Ce(NO_3)_3$	0.0090	$Rh(NO_3)_3$	0.0027
$Pr(NO_3)_3$	0.0032	$Pd(NO_3)_4$	0.0031
$Nd(NO_3)_3$	0.012	$Cr(NO_3)_3$	0.00013
$Eu(NO_3)_3$	0.0002	$Fe(NO_3)_3$	0.00057
$NaNO_3$	0.25	$Ni(NO_3)_2$	0.00006

Table 2

Kinetics of the extraction by 0.3M HDEHP + 0.15M TBP in n-alkanes from 0.5 M citric acid + 0.1 M NTA at pH 2.8

Shaking time, min	Distribution ratio					
	Am(III)	Eu(III)	Ce(III)	Sr(III)	Zr(IV)	Nb(V)
0.5	–	–	–	–	–	0.001
1	0.88	27	6.6	0.073	0.07	–
2	0.90	26	6.9	–	0.10	0.002
4	–	–	–	–	0.14	0.027
5	0.94	30	6.9	0.084	–	–
6	–	–	–	–	–	0.031
10	0.97	30	6.7	0.074	–	–
30	–	–	–	–	0.61	0.011
90	–	–	–	–	0.64	0.009

fraction of citric acid is bound in complexes with fission and corrosion products and, thus, at lower total citric acid concentrations the concentration of the free acid would be significantly influenced by changes in the composition of the waste solutions. As for improving the Am(III) extraction by rising the concentration of HDEHP beyond 0.3 M, this would make difficult the back extraction of Am(III) by the procedure discussed in Section II.

A small amount of a solid phase was formed during shaking at pH>2 in the lactic acid/NTA system and deposited at the interface after separation of the liquid phases. The solid contains Zr and Mo as the main components and Ba as a minor component, and its amount increases with increasing pH; nevertheless, at pH 2 - 3 it does not influence significantly the separation of the liquid phases after shaking.

The kinetics of the extraction in the presence of citric acid is illustrated by data gathered in Table 2. Zr(IV) and Nb(V) are extracted very slowly, while in the cases of americium and rare earths the extraction equilibrium is attained after 1 min shaking time.

The effect of TBP on the extraction of Am(III) and some fission products in the presence of citric acid is shown in Table 3 to be not pronounced at TBP concentrations between 0.05 and 0.3 M. Much more significant effects have been observed in extractions from simple aqueous solutions at very low loadings of the organic phase (6). The TBP concentration needed to prevent the formation of a third liquid phase in an alkaline washing of the spent organic phase is 0.15 - 0.2 M.

The influence of the degree of loading the organic phase in ex-
tractions from citrate solutions was studied by changing the
volume ratio of the phases. The results are given in Table 4.

The extractant dependencies from lactic and citric acid solutions
are shown in Figs. 3 and 4. The increase of the distribution
ratio with increasing HDEHP concentration is determined not only
by the composition of the extracted complexes, but also by the
increasing loading of the organic phase.

The effect of NTA on the extraction of Am(III), Eu(III), Zr(IV)
and Nb(V) is illustrated by Fig. 5. In a previous work (3) it
has been found that in extracting trace amounts of metals from
simple aqueous phases NTA improves markedly the separation of
Am(III) from Zr(IV). However, in this work with a complex aqueous
phase and a rather high loading of the organic phase the effect
of NTA on the separation of Am(III) from Zr(IV) is much less
pronounced. This can be due to a rather large fraction of NTA
bound in complexes with other fission and corrosion products.

2. Extraction from Noncomplexing Media

In these experiments the feed was prepared from a stock solution
of fission products containing only alkali, alkaline earth, and rare
earth elements as well as sodium nitrate. The concentrations of
the alkali, alkaline earth, and rare earth fission products in
the feed were as given in Table 1. The fission products were
chosen according to results of denitration experiments made in the
Waste Disposal Department (7). Thus in this set of experiments
conditions were simulated to correspond to concept No. 2.

pH Dependencies of the distribution ratios are shown in Figs. 6
and 7 for n-alkane diluent and in Fig. 8 for Solvesso-100, a
mixture of aromatic hydrocarbons. Fig. 6 illustrates the behaviour

Table 3

Effect of TBP concentration on the extraction by 0.3 M HDEHP/TBP-
/n-alkanes from 0.5 M citric acid + 0.1 M NTA at pH 2.8

Conc. of TBP M	Distribution ratio					
	Am(III)	Eu(III)	Ce(III)	Sr(II)	Zr(IV)	Nb(V)
0.00	1.03	33	6.0	0.054	0.0022	0.0014
0.05	0.97	27	5.8	0.053	0.0006	0.0016
0.1	0.97	28	5.4	0.060	0.0003	0.0013
0.3	0.95	26	5.5	0.076	0.0004	0.0010

Table 4

Effect of the phase volume ratio on the extraction by 0.3 M HDEHP
+ 0.15 M TBP in n-alkanes from 0.5 M citric acid + 0.1 M NTA at
pH 2.8 (volume of the aqueous phase 2.00 ml).

V_{org}	Distribution ratio			
	Am(III)	Eu(III)	Ce(III)	Sr(II)
2.5	0.86	24	5.5	0.098
4.0	-	38	12.5	0.32
5.0	1.70	33	13.1	0.29
6.0	1.81	38	15.2	0.34

of those elements the concentration of which is expected not to
be essentially lowered in the denitration of the waste solution.
The concentration of the elements listed in Fig. 7 is expected
to be reduced by a factor $\geqslant 100$ in the denitration. Thus only
radioactive tracers were added to the feed in the latter cases,
so that the concentration of these elements was $< 10^{-4}$M; at
this level they do not influence the degree of loading of
the organic phase and, consequently, also the distribution of
other extracted metals is not affected. Zr(IV) was distributed
in this run immediately after addition of the tracer to the
feed solution. If the aqueous feed is allowed to stand for some
time at pH 1 - 1.5, the nature of the Zr(IV) species may change
so that zirconium can become less extractable than shown in
Fig. 7. Hence, it is rather difficult to obtain consistent
results with Zr(IV) in the absence of a complexing agent in
the aqueous phase. Cr(III) and Ni(II) are not extracted; at
pH 0 - 1.6 their distribution ratios are $\leqslant 0.0013$ and $\leqslant 0.0006$
respectively.

As expected from previous studies (8), the efficiency of the
extraction of Am(III), Eu(III), Ce(III) and Sr(II) is higher
with the n-alkanes diluent than with Solvesso-100 by a factor of
about 10. As for the equilibrium pH region which is optimum for
extraction with HDEHP in n-alkanes, i.e. a pH between 1 and 1.5,
it may be difficult to hold such a pH value over a multistage
solvent extraction contactor. At the concentrations of extract-
able elements used throughout this work (see Table 1) the
liquid-liquid ion exchange reaction releases such amounts of
hydrogen ions to the aqueous phase that the equilibrium pH
becomes 0.8 - 1 even if the starting pH of the feed was 1.5 - 2
(this is in contrast to the extraction from complexed aqueous
solutions (Concept No. 1) where the complexant also has a
certain buffering action). In the batch experiments described
in this report pH values of >1 were attained by the addition of

NaOH to the feed; of course this caused precipitation of metal
hydroxides. Although the precipitate was dissolved during
the equilibration of the phases, this way of adjusting a suitable
equilibrium pH value is unacceptable for a countercurrent
extraction process. A possible manner of adjusting a pH between
1 and 1.5 in a countercurrent contactor could be a partial
neutralization of the HDEHP in the organic phase before extract-
ion. Table 5 shows how the equilibrium pH depends on the degree
of the HDEHP neutralization before shaking.

Table 5

Equilibrium pH of the aqueous phase as a function of the degree
of HDEHP neutralization in the starting organic phase. Starting
aqueous phase: see the left half of Table 1, pH ~2. Organic
TBP concentration 0.15 M.

Conc. of HDEHP M	Conc. of so- dium salt of HDEHP M	% sodium salt of HDEHP	Equilibrium pH
0.285	0.015	5	1.2
0.27	0.030	10	1.3
0.24	0.060	20	1.5
0.21	0.090	30	2.1
0.18	0.12	40	~3

The kinectics of the extraction (Fig. 9) is fast with Am(III),
Eu(III) and Ce(III) but significantly slower with multivalent
fission and corrosion products the bulk of which is expected
to be removed in the denitration step.

The extractant concentration dependence of the distribution ratios
of Am(III), Eu(III), Ce(III), and Sr(II) is shown in Fig. 10.

The effect of TBP on the extraction of Am(III), Eu(III) and
Ce(III) in the absence of complexing agents is more pronounced
(Fig. 11) than in their presence (Table 3). Surprisingly the
distribution of Eu(III) is affected more strongly than that
of Am(III) and Ce(III); this has not been observed in simple
systems with trace concentrations of the extracted metals (6).

The effect of Fe(III) on the extraction of Am(III), Eu(III)
and Ce(III) is illustrated by Fig. 12.

Loading of the organic phase with Mo(VI) does not affect
significantly the distribution ratios of Am(III), Eu(III),
and Ce(III) (Table 6).

II. Behaviour of Am(III) and Some Fission and Corrosion
    Products in the Partitioning Step

The partitioning step developed for Am/Cm isolation (1) is
based on the Talspeak process (9) utilizing diethylenetri-
aminepentaacetic acid (DTPA) as a selective complexant for
Am and Cm and may be described as a "Reverse-Talspeak" separat-
ion. Experiments were performed to obtain information directly
related to the particular conditions of the process under study
(rather high loading of the organic phase, use of n-alkanes
diluent, etc.) The dependencies of the distribution ratios of
Am(III), Ce(III), Eu(III), and Sr(II) on the pH and on the
DTPA concentration are shown in Figs. 13 and 14. Am(III) can
be separated from lanthanides(III) by selective back extraction
with 1 M lactic acid + 0.02 to 0.1 M DTPA from 0.3 M HDEHP +
0.15 M TBP in n-alkanes at pH 2.5 - 3.

The separation factors are somewhat lower than reported for
dodecane (9); this is most probably due to a higher loading
of the organic phase in this work. Crystallization of DTPA
from the aqueous phase can occur after several hours if the
pH is <3 and the DTPA concentration is >0.05 M. The difference
in the course of the pH dependence with Ce(III) and with

Table 6

Effect of Mo(VI) on the extraction by 0.3 M HDEHP + 0.15 M TBP in alkanes from a non-complexing aqueous phase

Conc.of Mo(VI) M	pH	Distribution ratio of Am(III)		D/D_o	pH	Distribution ratio of Ce(III)		D/D_o	pH	Distribution ratio of Eu(III)		D/D_o
		without Mo, D_o [a]	with Mo, D			without Mo, D_o [a]	with Mo, D			without Mo, D_o [a]	with Mo, D	
0.01	1.11	2.3	3.3	1.4	1.13	2.1	3.1	1.5	1.12	32	40	1.2_5
0.02	1.28	4.6	5.6	1.2	1.24	3.4	3.7	1.06	1.24	46	58	1.2_6
0.03	1.38	6.9	8.4	1.2	1.36	5.4	5.7	1.06	1.34	60	96	1.6

[a] Taken from the curves given in Fig. 6

Eu(III) and Am(III) can be ascribed to the lower stability
of the Ce(III)-DTPA complex in comparison with the Eu(III) and
Am(III) complexes (10-12).

The kinetics of the back extraction is illustrated by Fig. 15.
Obviously the choice of a suitably short phase contact time
can improve the separation of Am(III) from multivalent fission
and corrosion products left in the waste solutions after the
denitration step.

III. Behaviour of Some Fission and Corrosion Products in the
     Organic Phase Recovery Step

After the partitioning step the organic phase is still loaded
by lanthanides(III) and moreover , is contaminated by small
amounts of multivalent fission and corrosion products.
Lanthanides(III), namely Ce(III) and Eu(III) can be washed
out by 2 - 8 M HNO_3 (Fig. 16), while the washing of Y(III)
from the organic phase needs HNO_3 concentrations of $\geq$3M. No
successful back extraction of Fe(III) by nitric acid is possible
(Fig. 16), and the same can be expected with Zr(IV), RuNO(III)
and Pu(IV). For these elements an alkaline complexing solution
may be utilized for washing the organic phase. A sodium lactate
solution has been chosen for this purpose. Its concentration
of 0.5M is convenient: an effective washing of RuNO(III),
Zr(IV) + Nb(V), Fe(III) and Pu(IV) can be attained at a low
excess of NaOH, so that the total sodium ion concentration is
low enough for washing from the organic phase also mono-2-ethyl-
hexyl phosphate (which is expected to form in the radiation
decomposition of HDEHP). A 3 min phase contact is sufficient
for removing Zr(IV) + Nb(V), Fe(III), and Pu(IV) from the organic
phase, while the removal of RuNO(III) desires a contact time
of 20 min (Table 7).

Small amounts of Eu(III) and Y(III) remaining in the organic
phase after the back extraction by nitric acid are further
reduced by washing with sodium lactate + sodium hydroxide
solutions (Table 8). Surprisingly the remainder of cerium
is not washed out even after a 20 min contact time (Table 8);
as for the valence state of cerium under these conditions,
the oxidation to Ce(IV) cannot be excluded.

Table 7

Back extraction of multivalent fission and corrosion products by 0.5 M sodium lactate + 0-0.8M NaOH from 0.3 M NaDEHP + 0.2 M TBP

Conc. NaOH M	Distribution ratio						
	After 3 min shaking				After 20 min shaking		
	Zr(IV) + Nb(V)	Fe(III)	Pu(IV)	RuNO(III)	Pu(IV)	RuNO(III)	U(VI)
0.00	0.00020		0.0036	2.10	0.00111	0.20	0.0046
0.075	0.00019	0.0030	0.0045	2.02	0.0081	0.16	0.0037
0.20	0.00022	0.0094	0.0053	1.96	$0.017^{a)}$	0.13	$0.012^{a)}$
0.45	0.00025		$0.13^{a)}$	2.17	$0.021^{a)}$	0.10_5	$0.027^{a)}$
0.80	$0.0074^{a)}$	0.0054	$3.45^{a)}$	2.57	$0.054^{a)}$	0.20	$0.13^{a)}$

a) Loss of radioactivity from the liquid phases observed

Table 8

Back extraction of some trivalent fission products by 0.5 M sodium lactate + 0 - 0.8 M NaOH from 0.3 M NaDEHP + 0.2 M TBP

Conc. NaOH M	Distribution ratio			
	after 3 min shaking			after 20 min shaking
	Eu(III)	Y(III)	Ce	Ce
0.00	0.023	0.072	470	130
0.075	0.073	0.10	250	–
0.20	0.022	0.046	250	200
0.45	0.021	0.026	210	100
0.80	0.072	0.072	190	140

References

1) G. Koch, in A.S. Kertes and Y. Marcus (Eds.), Solvent Extraction Research, Wiley-Interscience, 1969, p. 349

2) S. Drobnik in H. Krause, Reports KFK-888 (1968) and KFK-1030 (1970)

3) Z. Kolarik, Proc.Int. Solvent Extraction Conference, The Hague, Holland, 19 - 23 April, 1971, in press

4) T. Moeller and R. Ferrus, Inorg.Chem. 1, 55 (1962)

5) S.H. Eberle und S.A.H. Ali, Radiochim. Acta 5, 58 (1966)

6) Z. Kolarik, J.Inorg.Nucl.Chem., in press

7) S. Drobnik, personnal communication

8) Z. Kolarik, Proc.Int.Conf.Solv.Extraction Chemistry, Gothenburg, Sweden (1966), p. 264, North-Holland, Amsterdam (1967)

9) B. Weaver and F.A. Kappelmann, Report ORNL-3559 (1964)

10) R.D. Baybarz, J.Inorg.Nucl.Chem. 27, 1831 (1965)

11) G. Anderegg, P. Nageli, F. Müller and G. Schwarzenbach, Helv.Chim. Acta 42, 827 (1959)

12) T. Moeller and L.C. Thomson, J.Inorg.Nucl.Chem. 24, 499 (1962)

F i g u r e s

Fig. 1: pH Dependence of the extraction by 0.3 M HDEHP + 0.15 M TBP in n-alkanes from 1 M lactic acid + 0.1 M NTA

Fig. 2: pH Dependence of the extraction by 0.3 M HDEHP + 0.15 M TBP in n-alkanes from 0.5 M citric acid + 0.1 M NTA (solid curves and empty points) and from 0.5 M citric acid + 0.05 M NTA (broken curves and solid points).

Fig. 3: Extractant concentration dependence in the extraction by 0.3 - 1.0 M HDEHP + 0.15 M TBP in n-alkanes from 1 M lactic acid + 0.1 M NTA at pH 2.1

Fig. 4: Extractant concentration dependence in the extraction by 0.3 - 1.0 M HDEHP + 0.15 M TBP in n-alkanes from 0.5 M citric acid + 0.1 M NTA at pH 2.2 (solid points and broken curves) and at pH 2.8 (empty points and solid curves).

Fig. 5: Effect of NTA on the extraction by 0.3 M HDEHP + 0.15 M TBP in n-alkanes from 1 M lactic acid + 0.01 - 0.1 M NTA at pH 2.2 (Am and Eu) and 1.7 (Zr and Nb)

Fig. 6: pH Dependence of the extraction by 0.3 M HDEHP + 0.15 M TBP in n-alkanes from a non-complexing aqueous phase.

Fig. 7: pH Dependence of the extraction by 0.3 M HDEHP + 0.15 M TBP in n-alkanes from a non-complexing aqueous phase.

Fig. 8: pH Dependence of the extraction by 0.3 M HDEHP in Solvesso-100 from a non-complexing aqueous phase.

Fig. 9: Shaking time dependence of the extraction by 0.3 M HDEHP + 0.15 M TBP in n-alkanes from a non-complexing aqueous phase.

Fig. 10: Extractant concentration dependence in the extraction by HDEHP + TBP in n-alkanes from a non-complexing aqueous phase. Concentration ratio HDEHP to TBP = 2.

Fig. 11: Effect of TBP on the extraction by 0.3 M HDEHP + 0 to
0.9 M TBP in n-alkanes from a non-complexing aqueous
phase.

Fig. 12: Effect of Fe(III) on the extraction by 0.3 M HDEHP +
0.15 M TBP in n-alkanes from a non-complexing aqueous
phase. Fe(III) is the aqueous concentration before
extraction.

Fig. 13: pH Dependence of the distribution between loaded 0.3 M
HDEHP + 0.15 M TBP in na-lkanes and 1 M lactic acid
+ 0.05 M DTPA. Starting loaded organic phase was
prepared by shaking with a 2/3 volume of the feed at
pH 1.

Fig. 14: Effect of DTPA on the distribution between loaded
0.3 M HDEHP + 0.15 M TBP in n-alkanes and 1 M lactic
acid + 0.005 to 0.1 M DTPA at pH 2.3. Starting loaded
organic phase was prepared by shaking with a 2/3
volume of the feed at pH 1.

Fig. 15: Shaking time dependence of the back extraction by 1 M
lactic acid + 0.05 M DTPA from loaded 0.3 M HDEHP +
0.15 M TBP in n-alkanes at pH 3.0. Starting loaded
organic phase was prepared by shaking with a 2/3
volume of the feed at pH 1.

Fig. 16: Effect of nitric acid on the distribution between
initially loaded 0.3 M HDEHP + 0.15 M TBP in n-alkanes
and 1 - 10 M HNO_3. Starting loaded organic phase was
prepared by shaking with a 2/3 volume of the feed at
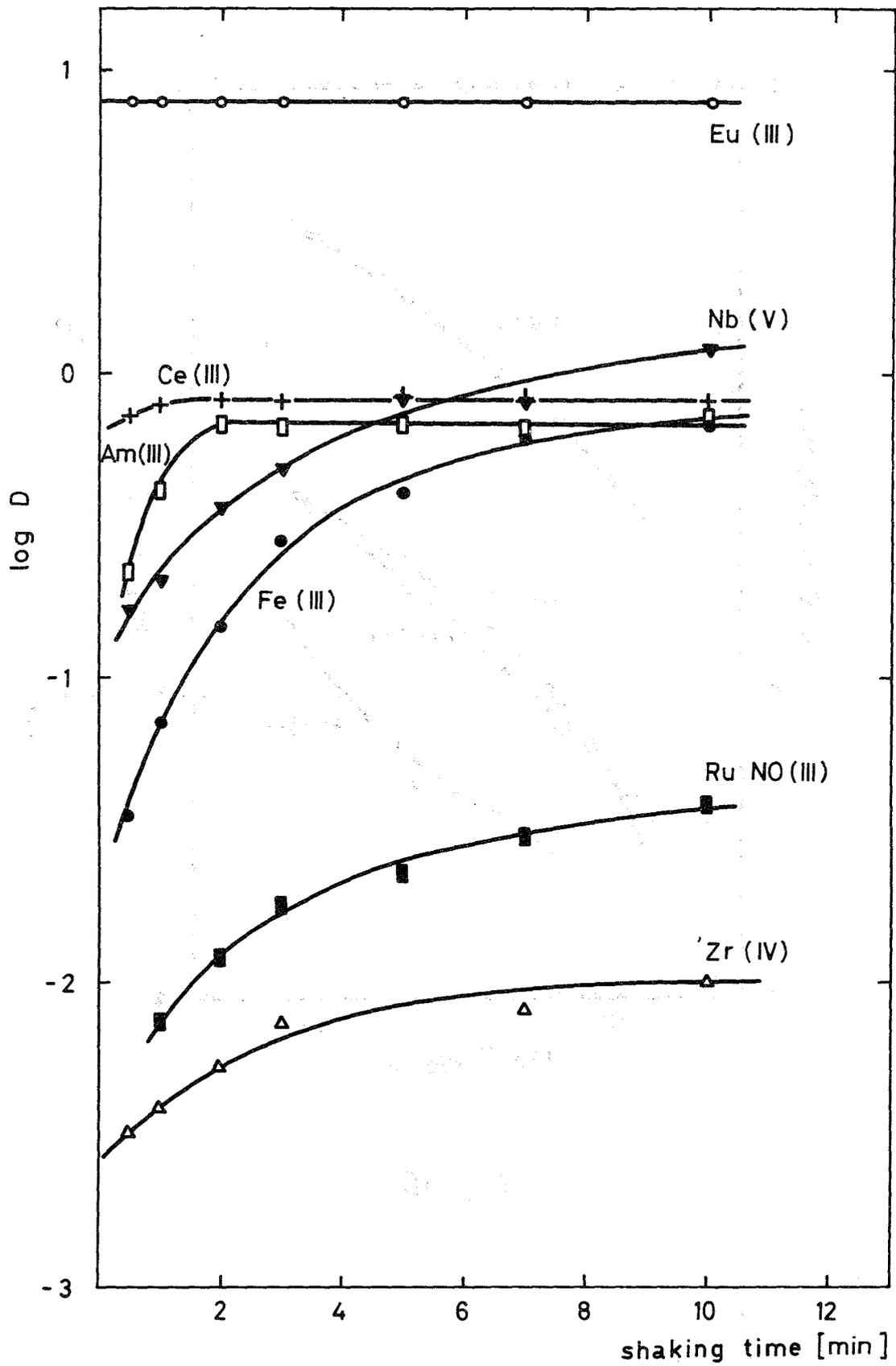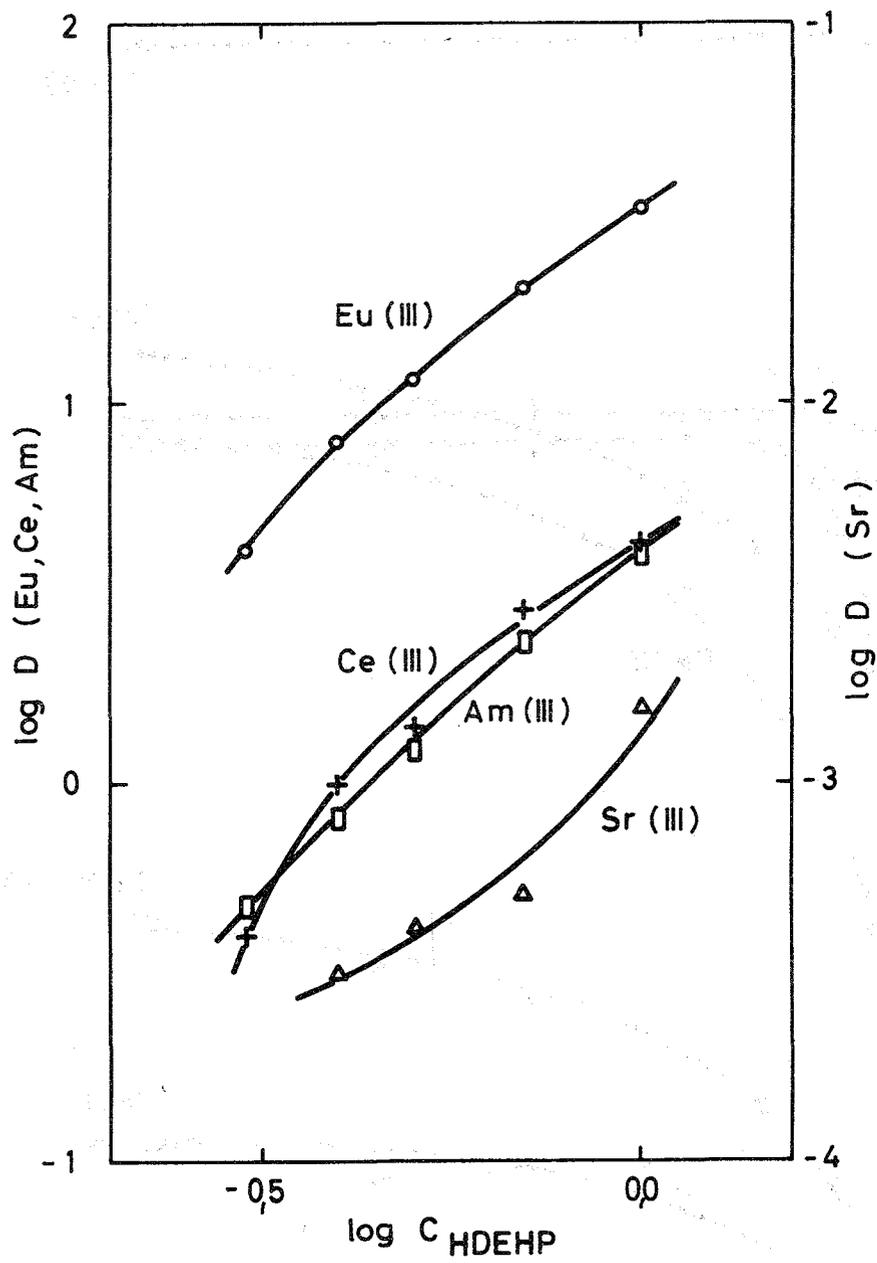pH 1.
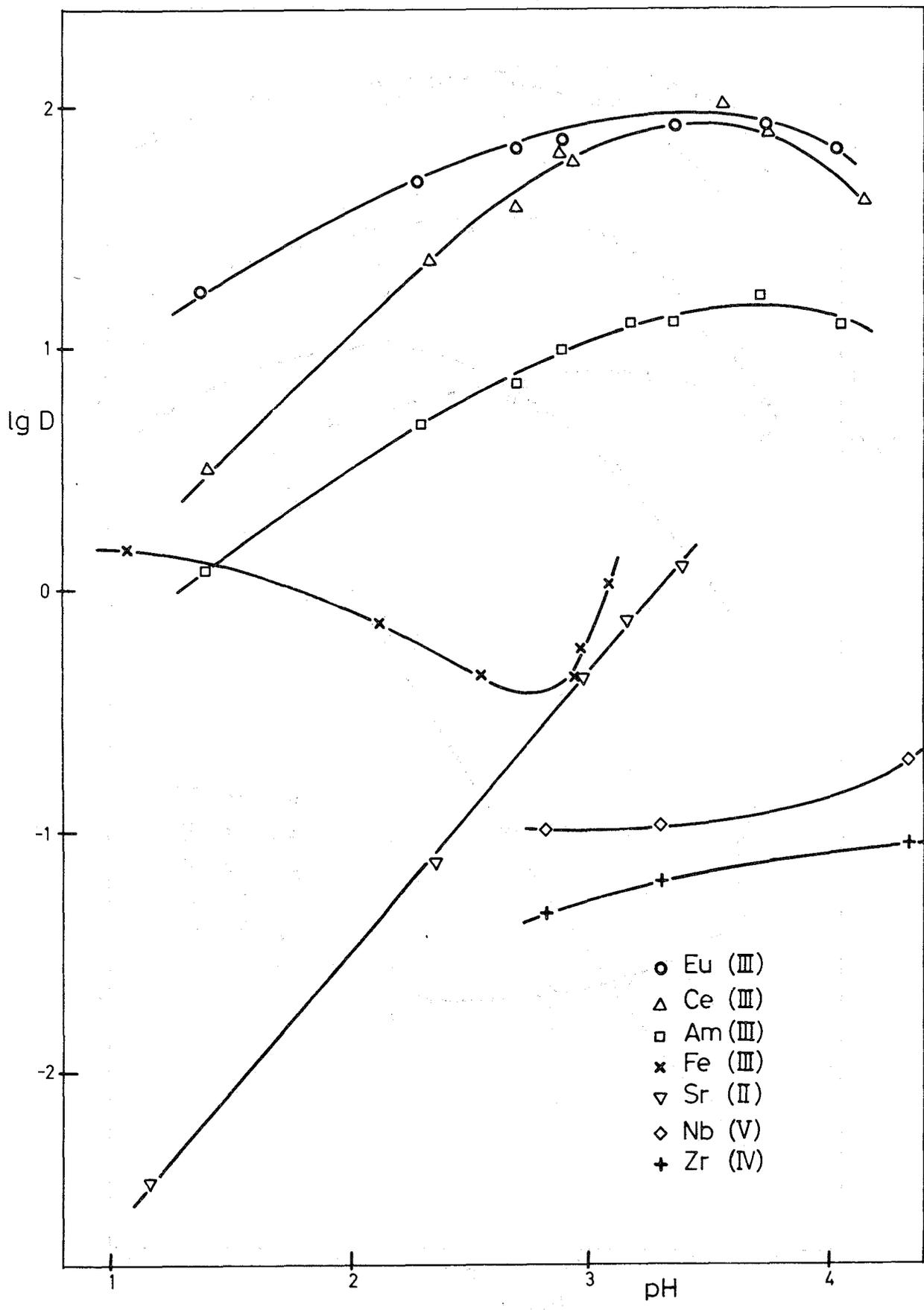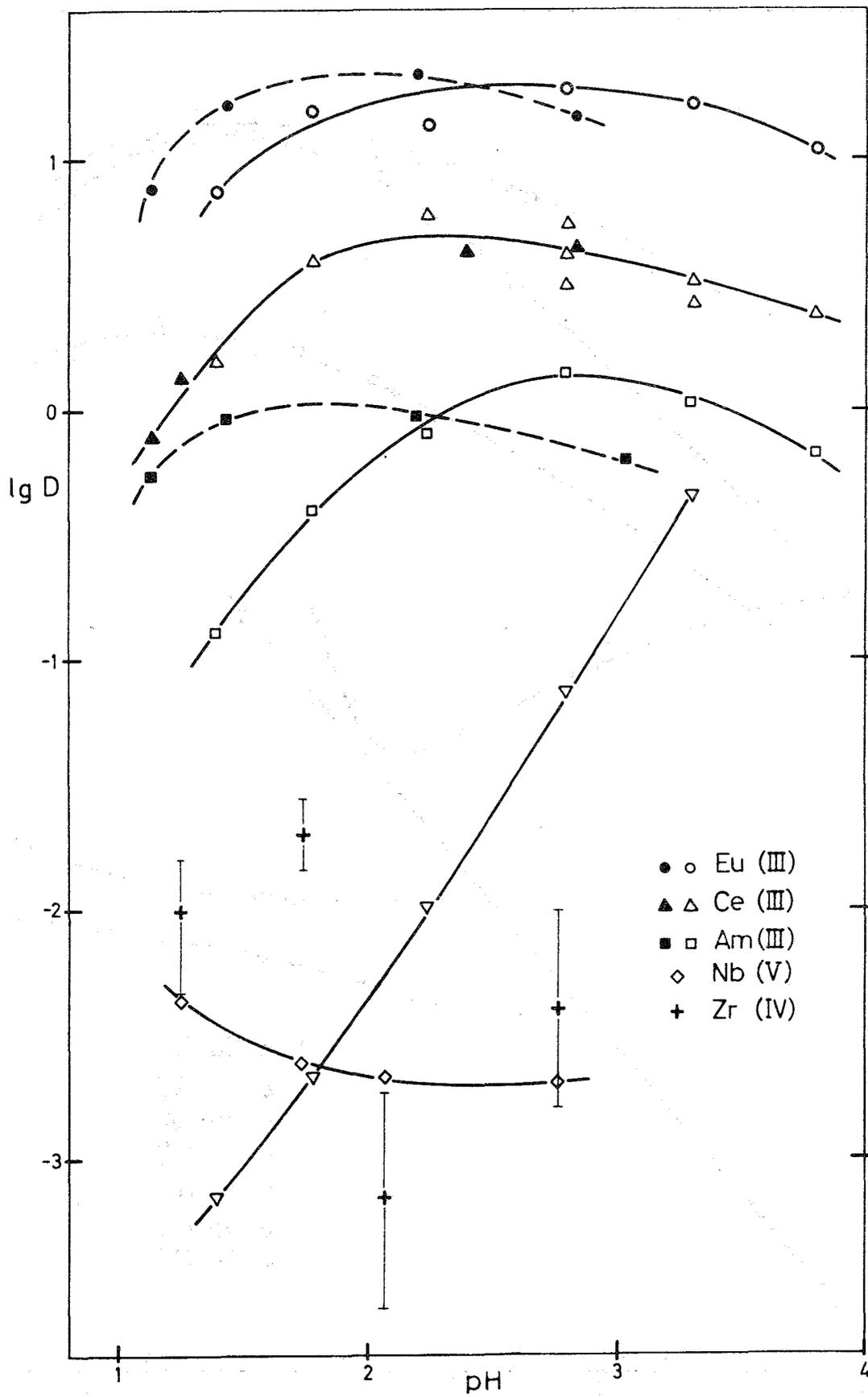
Fig. 1
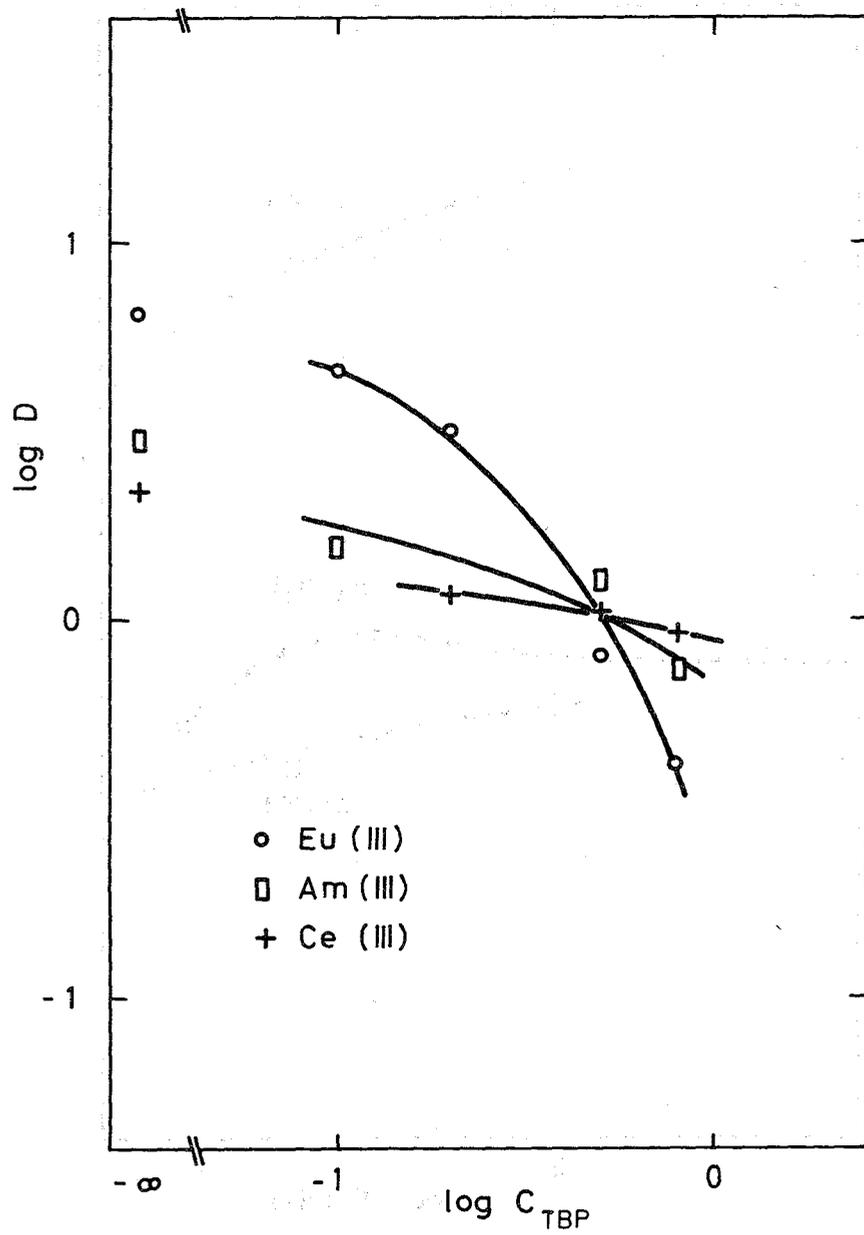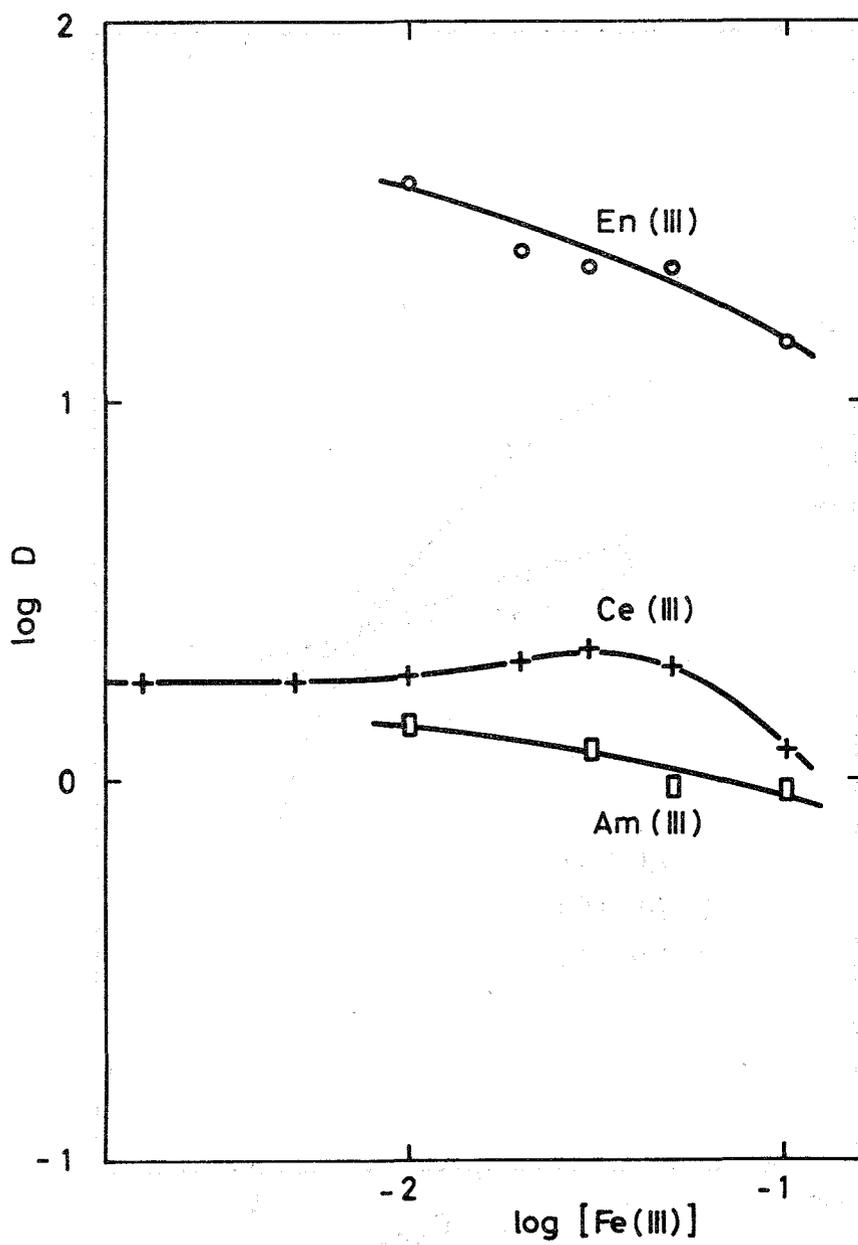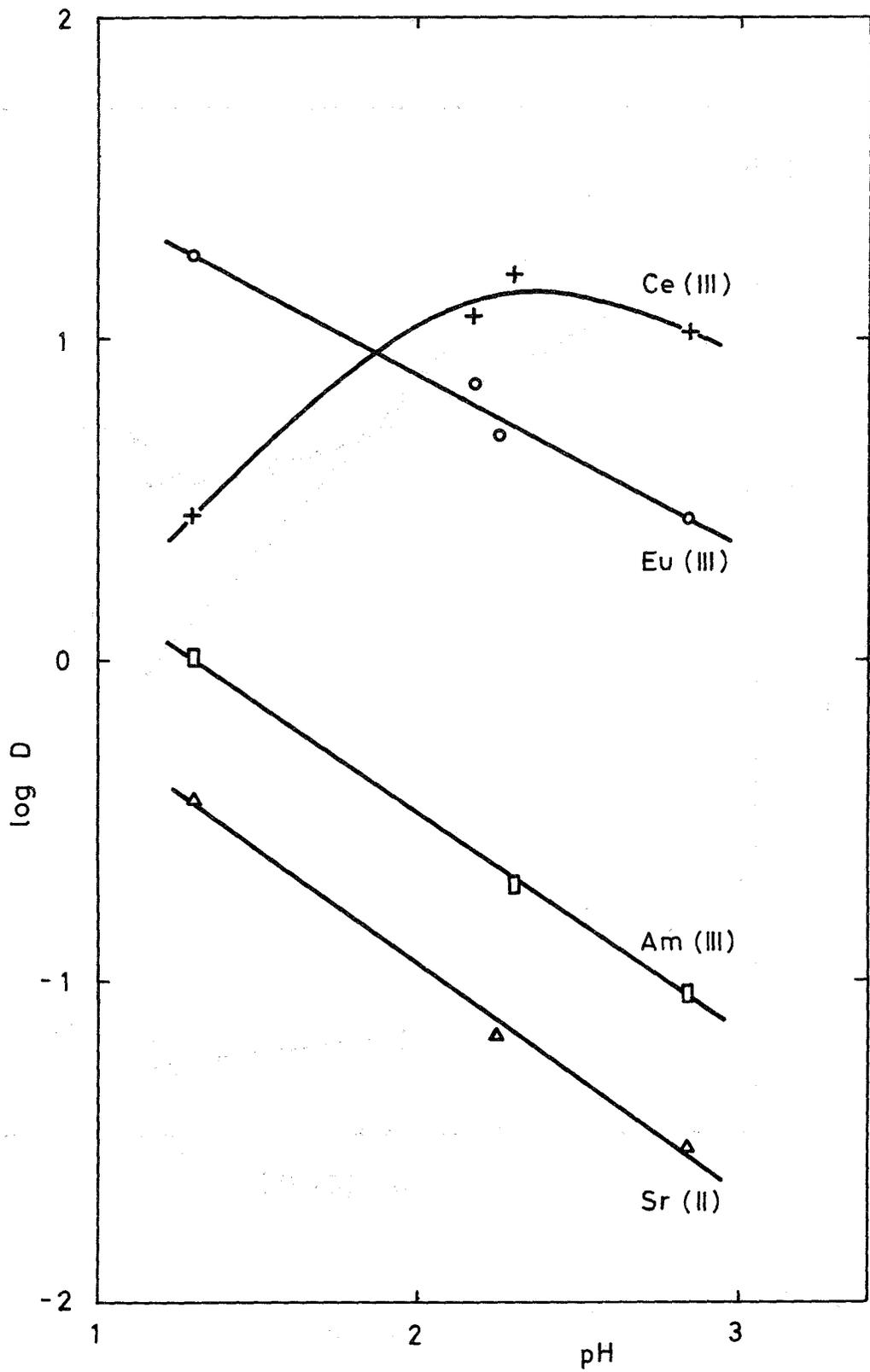
Fig. 2
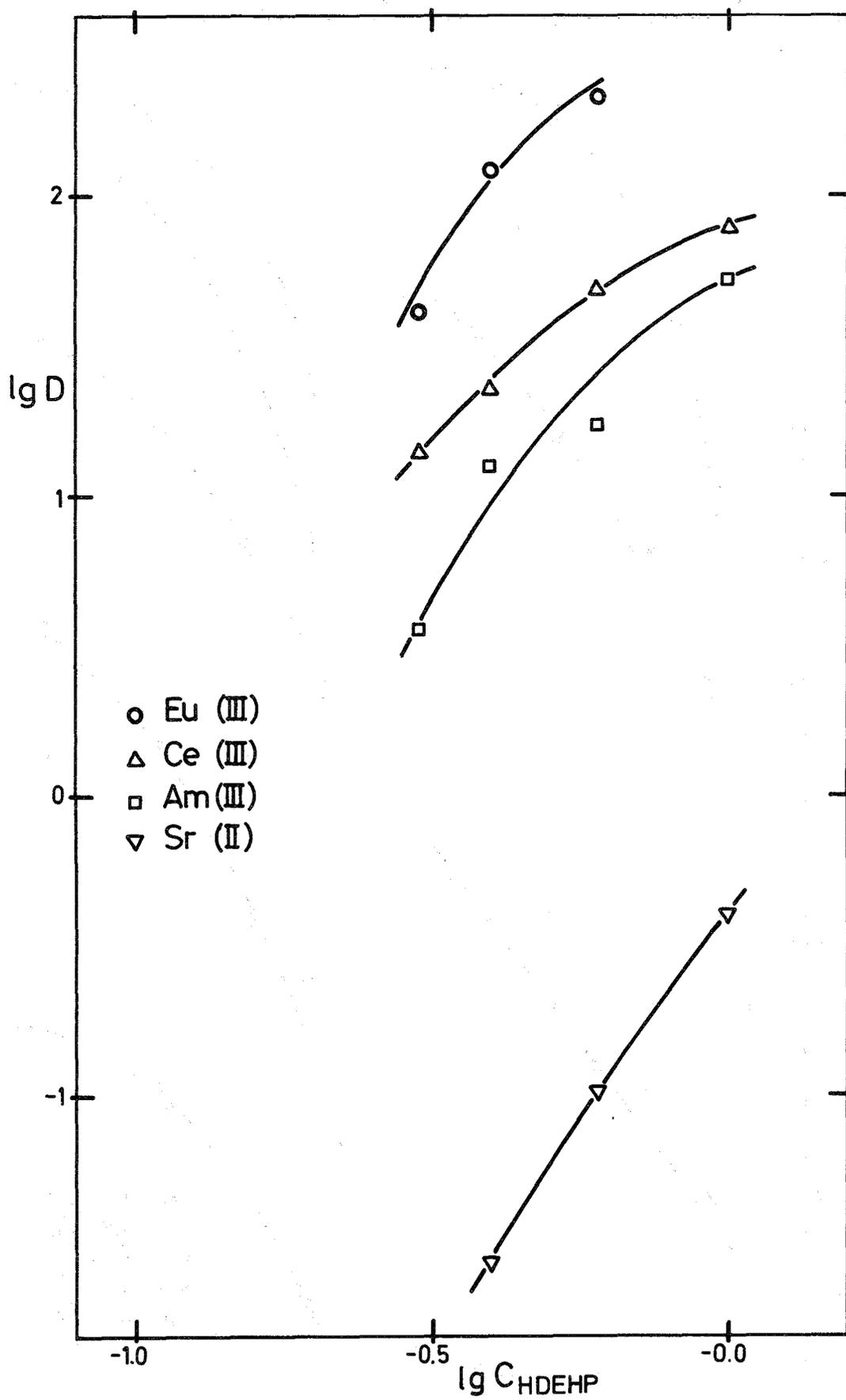
Fig. 3
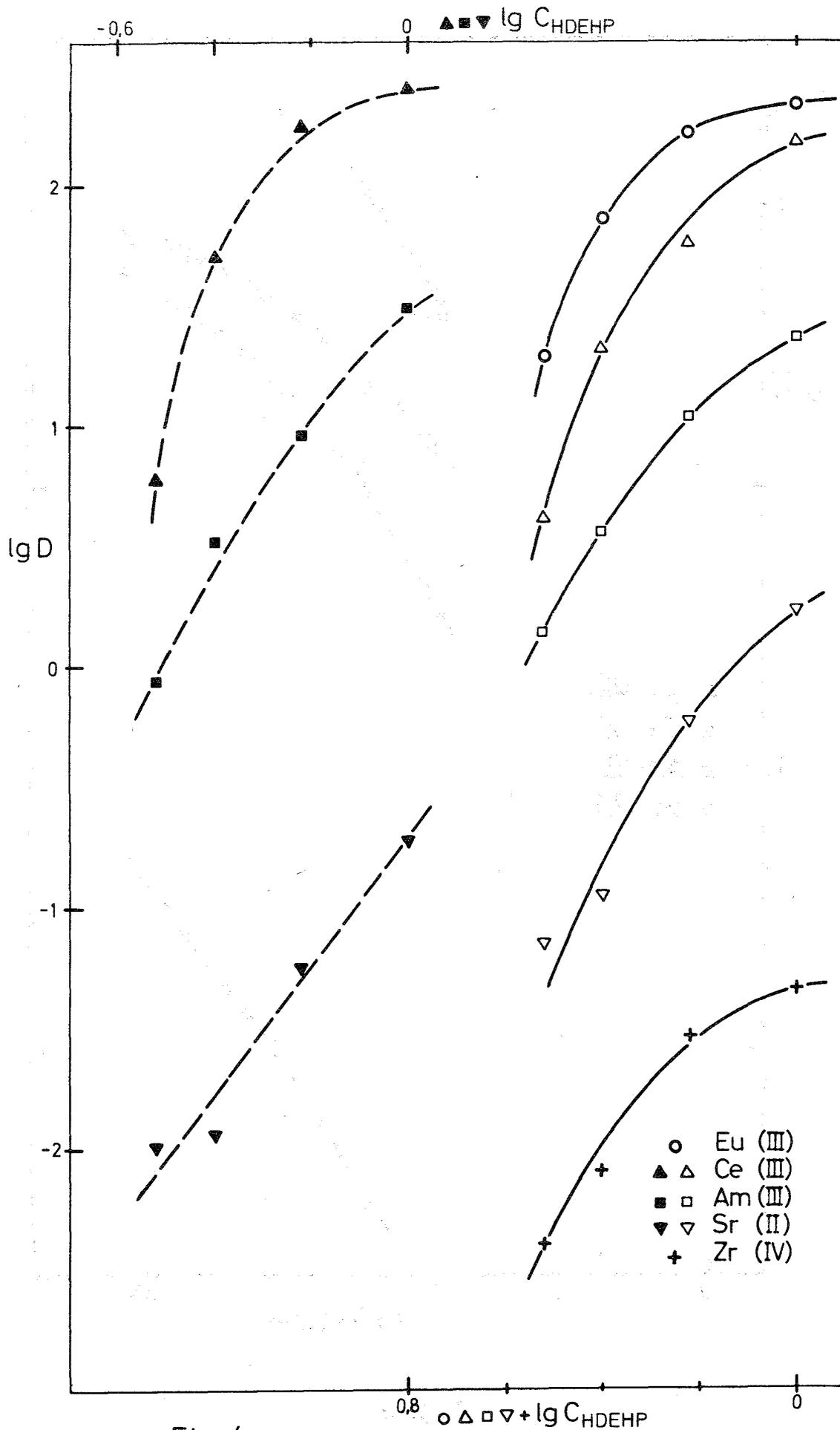
Fig. 4
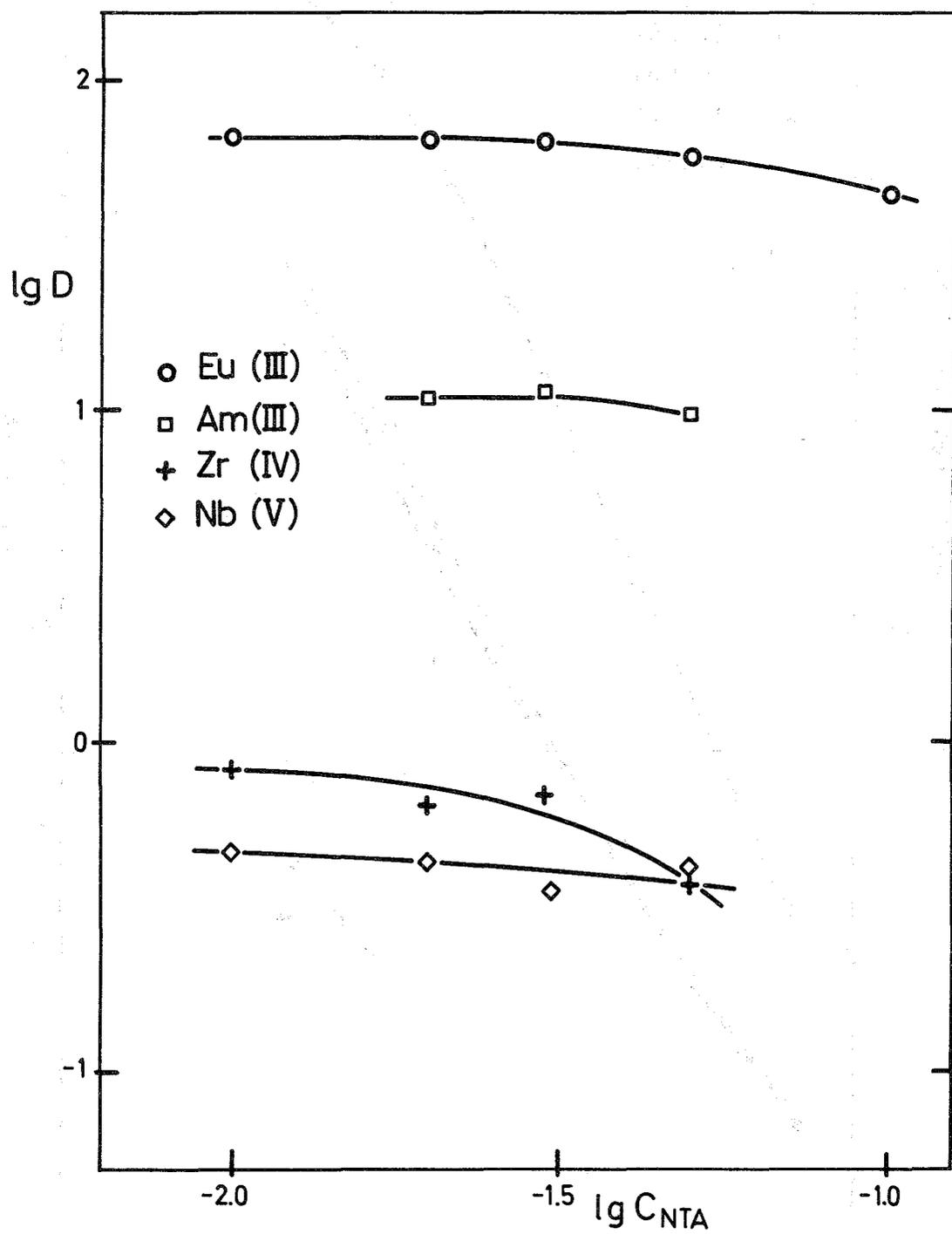
Fig. 5
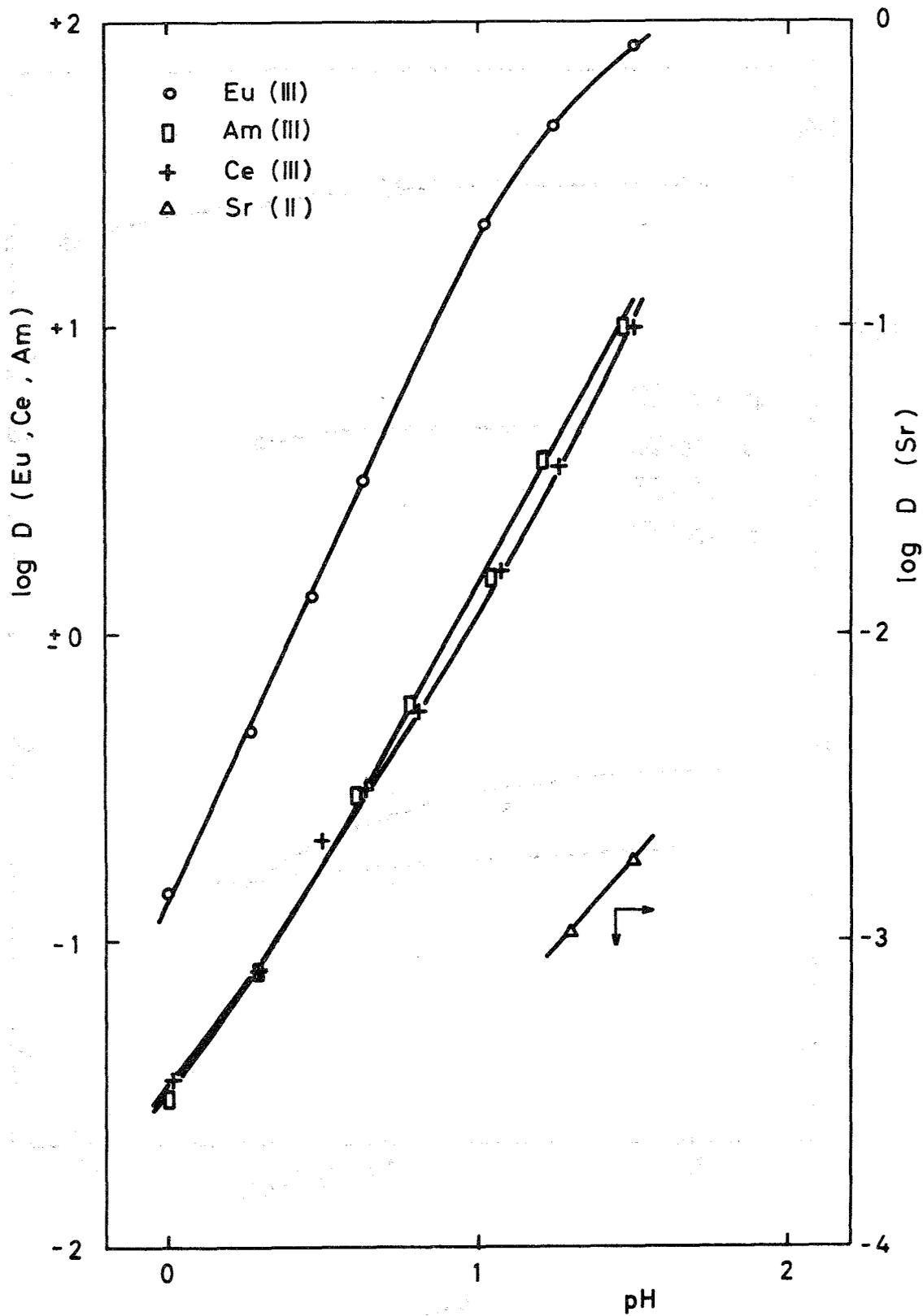
Fig. 6
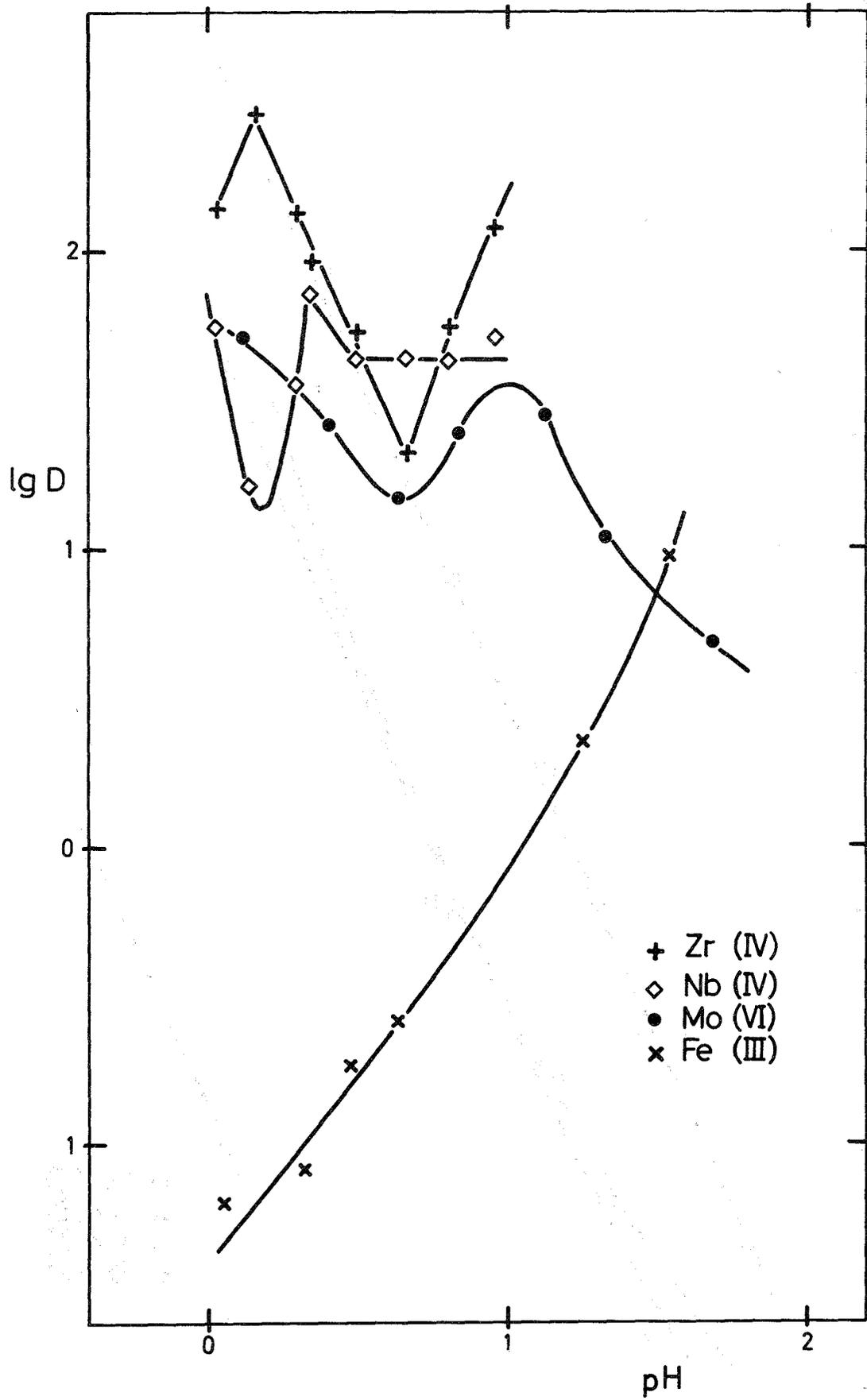
Fig. 7
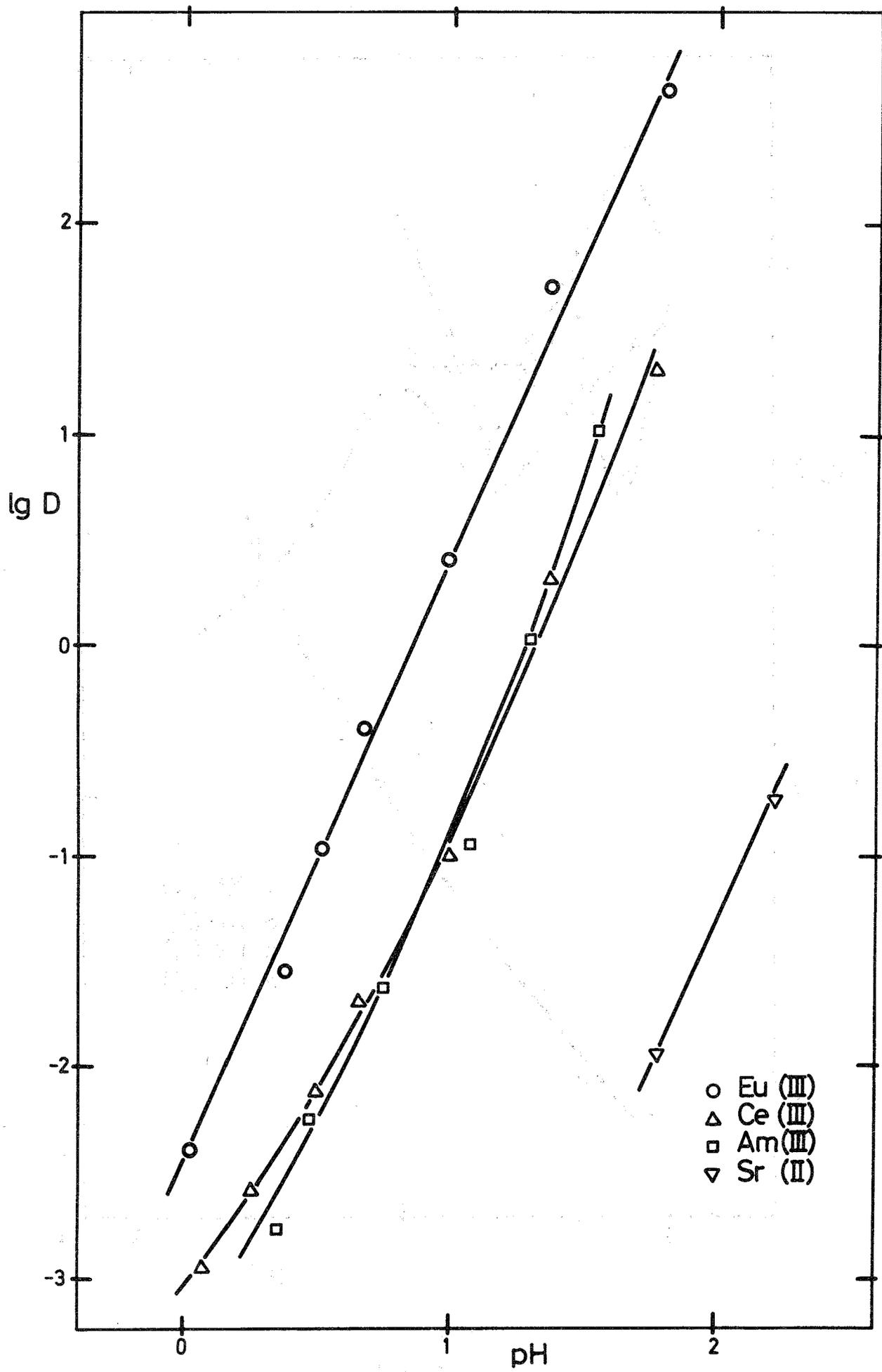
Fig. 8

Fig. 9

Fig. 10

Fig. 11

Fig. 12

Fig. 13

Fig. 14

Fig. 15

Fig. 16